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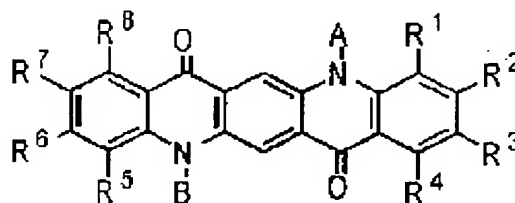
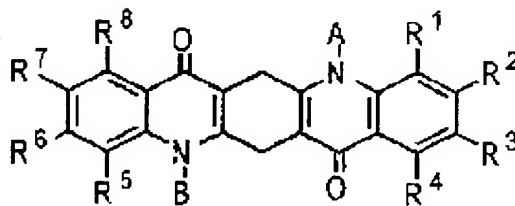
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## (54) FLUORESCENT COLORANT

## (57)Abstract:

PROBLEM TO BE SOLVED: To provide a fluorescent colorant having excellent heat resistance, solvent resistance, and light resistance, a plastic molded product comprising a masterbatch composed of the colorant and a resin, a coating material composition comprising a coating binder resin, and an ink composition composed of an ink binder.

SOLUTION: The fluorescent colorant is composed of a compound of formulae [1], [2] (wherein R1 to R8, A, and B are each independently, a hydrogen atom or a 1-50C organic residue, provided that at least four of R1 to R8, A, and B are the 1-50C organic residues).



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 CLAIMS
 

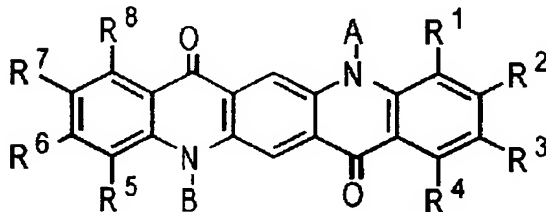
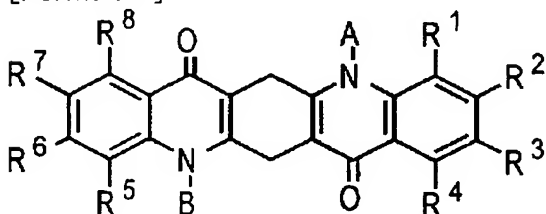
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[Claim(s)]

[Claim 1] The fluorescence coloring agent containing the compound shown by the following general formula [1].

General formula [1]

[Formula 1]



[0000] [-- among a formula, although R1-R8, and A and B express a hydrogen atom or the organic residue of carbon numbers 1-50 independently, respectively, at least four of R1-R8, and A and B are the organic residue of 1 to 50 as for them. ]

[Claim 2] The fluorescence coloring agent according to claim 1 with which the melting point of the compound shown by the general formula [1] is characterized by being 250 degrees C or more.

[Claim 3] The fluorescence coloring object which makes it come to contain a fluorescence coloring agent according to claim 1 in the resin of the shape of a particle which constructed the bridge.

[Claim 4] The fluorescence coloring plastic-molding object which makes it come to distribute a fluorescence coloring object according to claim 3 in plastics.

[Claim 5] The fluorescence coloring coating constituent containing a fluorescence coloring agent according to claim 1 and the binder for coatings.

[Claim 6] The fluorescence coloring ink constituent containing a fluorescence coloring agent and an ink binder according to claim 1.

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the fluorescence coloring agent used for various plastics moldingses, a coating, ink, etc. about a fluorescence coloring agent.

[0002]

[Description of the Prior Art] Generally fluorescent dye is rich in reactivity, and inferior to the robustness over light. As the cure, there are an approach of scouring this in a formation object, and the approach of coating-izing and painting as coloring agent powder which dissolved fluorescent dye in synthetic resin. These coloring agents should not yet be satisfied, although weatherability improves and it has come to be equal to field use, when fluorescent dye is scoured in a direct formation object.

[0003] On the other hand, the condensed multi-ring system organic compound which makes Quinacridone representation has lightfastness, thermal resistance, and the property excellent in solvent resistance, and the coloring agent of various coatings, such as a coating for automobiles, and polymeric materials, printing ink, etc. are utilized widely. These condensed multi-ring system organic compounds are firmly combined by the stacking or the intermolecular hydrogen bridge, consequently it does not dissolve in an organic solvent, or it is poorly soluble very much, and generally fluorescence is not seen or only very weak fluorescence is observed.

[0004]

[Problem(s) to be Solved by the Invention] Compatibility is high to a binder etc. and this invention aims at offering the fluorescence coloring agent excellent in lightfastness.

[0005]

[Means for Solving the Problem] It makes it easy for the fluorescence coloring agent shown in the general formula [1] to extend the intermolecular distance of a quinacridone pigment, and to distribute it by introducing four or more organic residue of carbon numbers 1-50 into the quinacridone pigment which was insoluble to the organic solvent until now, or its intermediate field. Consequently, when it was made the coating constituent which contains the binder for coatings when it is made the plastics moldings which it makes it come to distribute in plastics when making the fluorescence coloring agent of this invention contain in the resin of the shape of a particle which constructed the bridge, or when it was made the ink constituent containing an ink binder, pulling out the fluorescence which was not seen until now was found out.

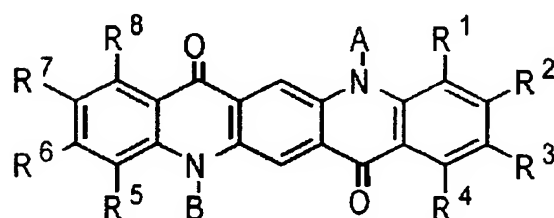
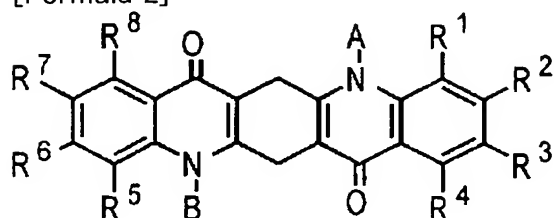
[0006] If concentration quenching explains this phenomenon, when the fused aromatic ring part of the Quinacridone molecule cannot approach mutually and will carry out it, molecule spacing is guessed for the phenomenon of breadth and quenching to stop being able to occur easily. When the fluorescence coloring agent shown in the general formula [1] introduced the organic residue of carbon numbers 1-50 into R1-R8, and A and B for the above reason, a stacking and an intermolecular hydrogen bridge are cut and it is expected to have become usable as a fluorescence coloring agent.

[0007] That is, this invention relates to the fluorescence coloring agent which consists of a compound shown by the following general formula [1].

General formula [1]

[0008]

[Formula 2]



[0009] [-- among a formula, although R1-R8, and A and B express a hydrogen atom or the organic residue of carbon numbers 1-50 independently, respectively, at least four of R1-R8, and A and B are the organic residue of 1 to 50 as for them.]

Furthermore, this invention relates to the above-mentioned fluorescence coloring agent with which the melting point of the compound shown by the general formula [1] is characterized by being 250 degrees C or more.

[0010] Furthermore, this invention relates to the fluorescence coloring object which makes it come to contain the above-mentioned fluorescence coloring agent in the resin of the shape of a particle which constructed the bridge.

[0011] Furthermore, this invention relates to the fluorescence coloring plastic-molding object which makes it come to distribute the above-mentioned fluorescence coloring object in plastics.

[0012] Furthermore, this invention relates to the fluorescence coloring coating constituent containing the above-mentioned fluorescence coloring agent and the binder for coatings.

[0013] Furthermore, this invention relates to the fluorescence coloring ink constituent containing the above-mentioned fluorescence coloring agent and an ink binder.

[0014]

[Embodiment of the Invention] R1-R8 of the compound shown by the general formula [1] in this invention, and A and B are a hydrogen atom or the organic residue of carbon numbers 1-50, and the aryl thio radical which is not permuted [ the aryloxy group which is not permuted / the aryl group which is not permuted / the thio alkoxy group which is not permuted / the alkoxy group which be permuted / the alkyl group which be permuted / a permutation or / a permutation, or / a permutation, or / a permutation, or /, a permutation, or /, a permutation, or ] is mentioned as an example of the organic residue of carbon numbers 1-50.

[0015] As an alkyl group which is not permuted [ a permutation or ], a methyl group, an ethyl group, n-propyl group, An ISO-propyl group, butyl, sec-butyl, tert-butyl, A pentyl radical, a hexyl group, a heptyl radical, an octyl radical, the dodecyl, a tetradecyl radical, An EOSHIRU radical, a heptadecyl radical, a hexadecyl radical, a cyclohexyl radical, Others [ alkyl group / of the carbon numbers 1-50, such as an octadecyl radical and a stearyl radical, / non-permuted ], 2-phenyl isobutyl radical, a trifluoro pentyl radical, a hexafluoro pentyl radical, There is a permutation alkyl group of the carbon numbers 1-50, such as benzyl, alpha-phenoxy benzyl, alpha, and alpha-dimethylbenzyl radical, alpha, and alpha-methylphenyl benzyl, alpha, and alpha-ditrifluoromethyl benzyl, a triphenylmethyl radical, and alpha-benzyloxybenzyl radical.

[0016] As an alkoxy group which is not permuted [ a permutation or ] Others [ alkoxy group / of the carbon numbers 1-50, such as an n-butoxy radical, a t-butoxy radical, n-octyloxy radical, and t-octyloxy radical, / non-permuted ], There are permutation alkoxy groups of carbon numbers 1-50, such as 1, 1, and 1-tetrafluoro butoxy radical and a benzyloxy radical. As a thio

alkoxyl group which is not permuted [ a permutation or ] A thio n-butoxy radical, a thio-t-butoxy radical, a thio-n-octyloxy radical, There are permutation thio alkoxy groups of carbon numbers 1-50, such as 1 and 1, and 1-tetrafluoro thio butoxy radical and a benzyloxy radical. [ besides the non-permuted thio alkoxy group of the carbon numbers 1-50, such as a thio-t-octyloxy radical, ]

[0017] As an aryl group which is not permuted [ a permutation or ], there are a phenyl group, 2-methylphenyl radical, 3-methylphenyl radical, 4-methylphenyl radical, 4-ethyl phenyl group, a biphenyl radical, 4-methyl biphenyl radical, 4-ethyl biphenyl radical, 4-cyclohexyl biphenyl radical, a terphenyl radical, 3, 5-dichlorophenyl radical, a naphthyl group, 5-methyl naphthyl group, an anthryl radical, a pyrenyl radical, etc. As an aryloxy group which is not permuted [ a permutation or ], there are permutation aryloxy groups of carbon numbers 1-50, such as 1 and 1 besides the non-permuted aryloxy group of the carbon numbers 1-50, such as a phenoxy group, 4-ethyl phenoxy group, 4-propyl phenoxy group, a 4-n-butyl phenoxy group, a 4-t-butoxy phenoxy group, a 2-n-octyl phenoxy group, and a 3-t-octyl phenoxy group, 1-tetrafluoro phenoxy group, and a biphenyl oxy-radical.

[0018] As an aryl thio radical which is not permuted [ a permutation or ], there are a phenylthio radical, 2-methyl phenylthio radical, 3-methyl phenylthio radical, 4-methyl phenylthio radical, 4-ethyl phenylthio radical, a biphenyl thio radical, 4-methyl biphenyl thio radical, 4-ethyl biphenyl thio radical, 4-cyclohexyl biphenyl thio radical, a TAFENIOCHIO radical, 3, 5-dichloro phenylthio radical, a naphthyl thio radical, 5-methyl naphthyl thio radical, an anthryl thio radical, a pyrenyl thio radical, etc.

[0019] As an example of the substituent which organic residue may combine, there is an amino group other than the above-mentioned functional group which is not permuted [ a halogen atom, a permutation, or ].

[0020] There are fluorine, chlorine, a bromine, and iodine as a halogen atom, and there are a phenyl methylamino radical besides dialkyl permutation amino groups, such as an amino group, a dimethylamino radical, and a diethylamino radical, a diphenylamino radical, a ditolylamino radical, a dibenzylamino radical, etc. as an amino group which is not permuted [ a permutation or ].

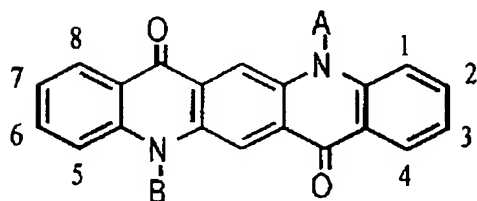
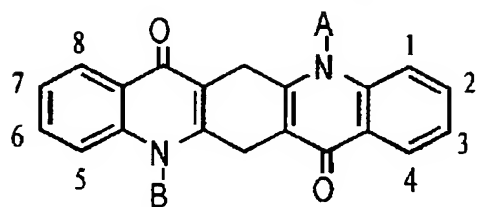
[0021] Moreover, by adjoining substituents, it may join together mutually, respectively and a phenyl group, a naphthyl group, an anthryl radical, a pyrenyl radical, etc. may be formed.

[0022] Especially the location that the organic residue of the compound of the general formula [1] in this invention may combine is not limited. However, it is desirable preferably that it is the 1.4.5.8th place shown in the general formula [2] or that A and B are organic residue, and when A or B is an aryl group, it is still more desirable that 1 or the organic big residue of steric hindrance is introduced the 5th place. It is thought that such organic residue serves to cut or pull apart an intermolecular hydrogen bridge.

General formula [2]

[0023]

[Formula 3]



[0024] [-- A and B express a hydrogen atom or the organic residue of carbon numbers 1-50 independently among a formula, respectively.]

generally, a fluorescent pigment or fluorescent dye has the low melting point, and it is fluorescent dye of versatility -- acid When it is red, it is 208 degrees C. When the melting point of a fluorescent pigment was low and it is made a masterbatch, the problem of discoloration, metal mold dirt, etc. is caused. In order to avoid these problems, the melting point of 250 degrees C or more is required at worst.

[0025] N,N-dibenzyl-1,5-dimethyl-Quinacridone which is the compound shown by the general formula [1]: The melting point of a compound (1) is as high as 350 degrees C, and it has strong fluorescence. The compound shown by the general formula [1] is compoundable by compounding Quinacridone using the raw material which introduces organic residue into a commercial quinacridone pigment and which is caused especially or has organic residue. The example of the compound shown by the general formula [1] below is illustrated to Table 1.

[0026]

[Table 1]

化合物	化学構造
(1)	
(2)	
(3)	

[0027]

化合物	化学構造
(4)	
(5)	
(6)	

[0028]

化合物	化学構造
(7)	
(8)	
(9)	

[0029]

化合物	化学構造
(10)	
(11)	
(12)	

[0030]

化合物	化学構造
(13)	
(14)	
(15)	

[0031]

化合物	化学構造
(16)	
(17)	
(18)	

[0032] In a solid state, it has strong fluorescence and the compound shown by the general formula [1] shows the high melting point. For this reason, although the compound shown by the general formula [1] can be used as a fluorescence coloring agent as it is, after distributing the compound shown by the general formula [1] in the resin of cross-linking, it can be made to be able to harden and what ground this to the particle can also be used as a fluorescence coloring agent. Moreover, after emulsion-izing the compound shown by the general formula [1] by the drainage system with aqueous resin using an emulsifier or a stabilizer, what was hardened and was made into the spherical particle can be used as a fluorescence coloring agent. As resin of the above-mentioned cross-linking, acrylic resin, styrene resin, aromatic series sulfonamide resin, an epoxy resin, an alkyd resin, a urea resin, melamine resin, benzoguanamine resin, acrylic urethane resin and those copolymers, copolycondensation resin, etc. can be mentioned. An ultraviolet ray absorbent, an anti-oxidant, a singlet oxygen quencher, a hindered amine light stabiliser, other stabilizers, and an additive can be blended with the above-mentioned resin.

[0033] 0.1-100 micrometers of mean particle diameter of said particle are 1-20 micrometers preferably. The concentration of the compound shown by the general formula [1] blended with the resin of a particle is 0.05 - 10 % of the weight preferably 0.01 to 20% of the weight. It becomes [ fluorescence intensity ] weak that it is low concentration and is not more desirable than 0.01 % of the weight.

[0034] As a more concrete manufacturing method of the fluorescence coloring agent which consists of particle-like resin For example, after adjusting the mixture of amino compounds, such as benzoguanamine and a melamine, and formalin so that pH may serve as the range of 5-10, In the primary reaction object of the amino resin which was made to react at the temperature of 50-100 degrees C, and was obtained The amino resin 100 weight section is received in the compound shown by the general formula [1]. 0.1 - 20 weight section, Under stirring in the water solution which carries out 0.5-10 weight section addition preferably, and contains protective colloid agents, such as polyvinyl alcohol, for this in the range of 1 - 30 weight section to the amino resin 100 weight section, It supplies and the suspension of amino resin is obtained, and subsequently, in addition, 40-100 degrees C performs polycondensation hardening for curing catalysts, such as a mineral acid and an organic acid, at this to the amino resin 100 weight section in the range of 0.01 - 10 weight section, and after it carries out the obtained hardening resin a \*\* exception and it carries out stoving, it crushes.

[0035] The fluorescence coloring agent which consists of the compound shown by the general formula [1] or a compound shown by the general formula [1], and bridge formation resin of a particle can be blended with shaping plastics. Under the present circumstances, the fluorescence coloring agent of this invention can be blended with high concentration, for example, extrusion molding can be carried out to the constituent which consists of dispersants, such as base resin and metal soap, and a wax, at a pellet type, and it can consider as a masterbatch. As resin of the tree for shaping plastics, there are polyolefin resin, such as polyethylene resin and polypropylene resin, polyester resin, polyamide resin, PVC resin, ABS plastics, styrene resin, acrylic resin, polycarbonate resin, urethane resin, amino resin, etc. Let these thermoplastics be a plastics moldings by the approach of inflation molding, calender shaping, and others. The fluorescence coloring agent of this invention can be blended with thermosetting plastic, such as an epoxy resin, phenol resin, polyester resin, polyester imide resin, and polyamidoimide resin.

[0036] Surfactants, such as rosin acid soap, stearin acid soap, oleic acid soap, Na-G beta-naphthyl methane JISARUFATO, Na-lauryl sulfate, Na-diethyl hexyl sulfo KUSHINETO, and Na-dioctyl sulfo KUSHINETO, can distribute, and the fluorescence coloring agent of this invention can be used as the coloring agent constituents a coating, for ink, etc.

[0037] The compound shown by the general formula [1] is meltable to an organic solvent, and can blend this with a coating or ink as remaining as it is or a fluorescence coloring agent of solvent insolubility which consists of a particle of resin. As ink, there are printing ink, such as offset-printing ink, gravure ink, and the silk screen, ink for ink jets, copying ink, etc. As paint resin and resin for ink, polymethyl acrylate, Acrylic resin, such as polyethylacrylate, polymethylmethacrylate, and polyvinyl acrylate, Polyurethane resin, acrylic urethane resin, polyester resin, polyarylate resin, Vinyl chloride resin, an ethylene-vinylacetate copolymer,

polyvinyl formal resin, Amino resin, an alkyd resin, an epoxy resin, phenol resin, polyester resin, Polyester imide resin, polyamidoimide resin, silicone resin, Polyvinylidene chloride resin, polyethylene vinyl alcohol resin, polyolefin resin, Elasticity, such as vinylidene chloride resin, butadiene resins, styrene resin, phenoxy resin, polyamide resin, cellophane, ethyl cellulose, and a nitrocellulose, rigid resin, etc. can be mentioned. The blending ratio of coal of the fluorescence coloring agent of this invention is 0.5 - 10 % of the weight preferably 0.1 to 20% of the weight to a coating constituent or the whole ink constituent.

[0038] The fluorescence coloring agent of this invention can raise light stability-proof by using it with an ultraviolet ray absorbent. An ultraviolet ray absorbent An o-hydroxy benzophenone, 2-hydroxy-4-n-octoxybenzophenone, Benzophenone systems, such as 2-hydroxy-4-methoxybenzophenone, 2-(2'-hydroxyphenyl) benzotriazol, 2-(2'-hydroxy-5'-t-octyl phenyl) benzotriazol, 2-(2'-hydroxy-3'-t-butyl -5'-methylphenyl)-5-chlorobenzo triazole, Benzotriazols, such as 2-(2'-hydroxy-5'-methylphenyl) benzotriazol, Ethyl-2-cyano - 3 and 3-diphenyl acrylate, 5-ethylhexyl-2-cyano - Cyanoacrylate systems, such as 3 and 3-diphenyl acrylate, Salicylic acid systems, such as phenyl salicylate and 4-t-butyl phenyl salicylate, Ultrafine particle inorganic systems, such as oxalic acid anilide systems, such as 2-ethyl-5'-t-butyl-2'-ethoxy-N and an N'-JIFENIROKISARU amide, a zinc oxide, titanium oxide, and a zirconium dioxide, etc. can be used.

[0039]

[Example] Hereafter, this invention is further explained to a detail based on an example. The section expresses the weight section among an example.

The synthetic example 1 (composition of a compound (1))

Into the dioxane 300 section, the succinyl succinic-acid dimethyl 24 section, the 2-amino-1-methylbenzene 50 section, and the 35% hydrochloric-acid 1.5 section were put in, and heating churning was carried out at 95 degrees C for 3 hours. Then, it diluted with the methanol water of the 500 sections, and the rare sodium-hydroxide water solution neutralized. The depositing yellow solid-state was carried out the \*\*\*\*\* exception, and it washed in the methanol 500 section further. The obtained yellow solid-state was dried at 70 degrees C, and the needle crystal 43 section which has yellow fluorescence was obtained.

[0040] Heating stirring was performed for this yellow needle crystal 50 section at 250 degrees C under the nitrogen air current with the 1-methylnaphthalene 300 section for 10 hours. Then, the orange solid-state which added the methanol 300 section and deposited was carried out the \*\*\*\*\* exception. It washed in the methanol 300 section, the obtained orange solid-state was dried at 70 degrees C, and the light orange powder 28 section which has whitish and orange fluorescence was obtained.

[0041] Heating stirring was performed for this light orange crystal 20 section for 10 minutes at 280 degrees C under the air air current with the dibutyl phtalate 200 section. Then, the purple solid-state which added the methanol 300 section and deposited was carried out the \*\*\*\*\* exception. It washed in the methanol 300 section, the obtained purple solid-state was dried at 70 degrees C, and the purple powder 18 section which has orange fluorescence was obtained. Heating stirring was performed for this purple powder 18 section at 100 degrees C for 10 hours with the dichlorobenzene 400 section, the 50% caustic soda water-solution 1000 section, the benzyl triethyl ammonium chloride 9 section, and the benzyl bromide 160 section. Then, the chloroform 300 section was added and the resultant was extracted. This chloroform layer was washed with water until it became neutrality, and it condensed after desiccation, the column chromatography using silica gel refined, and the powder 8 section which reprecipitates with a methanol and has orange fluorescence was obtained. It checked that it was a compound (1) as a result of molecular weight analysis. The fluorescence spectrum of a product is shown in drawing 1 . In addition, the melting point of this compound was 350 degrees C.

The synthetic example 2 (composition of a compound (3))

Heating churning of 1, the 5-dimethyl Quinacridone 8 section, the benzene 140 section, the 50% caustic soda water-solution 500 section, the benzyl triethyl ammonium chloride 6 section, and the alpha-BUROMO-O-xylene 50 section was violently carried out at 80 degrees C among the flask for 30 hours. Then, the decantation separated the solid-state part, the water of the 500

sections washed, and chloroform extracted. This chloroform layer was condensed and the solid-state 8 red section was obtained. The column chromatography using silica gel refined and the powder 3 section which reprecipitates with a methanol and has yellow fluorescence was obtained. It checked that it was a compound (3) as a result of molecular weight analysis. The fluorescence spectrum of a product is shown in drawing 2. In addition, this compound showed the melting point of 320 degrees C.

The powder 30 section of the compound (1) obtained in the example 1 of example 1 composition, polyethylene (trade name "SUMIKASEN G-808" by Sumitomo Chemical Co., Ltd. 30 section, and polyethylene wax (the trade name by Mitsuhiro chemical-industry company "Sun Wacks 131P" 40 section was pelletized after kneading and with an extruder by the kneader, and the master batch was obtained.) It is high density polyethylene (it mixed with the trade name "high ZEKKUSU 2208" by Mitsui Petrochemical Industries, Ltd. 100 section, and the resin moldings was obtained by extrusion molding.) about this masterbatch 4 section. As a result of observing change of fading after 72-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted. The same actuation as an example 1 was repeated except having changed the compound (1) in example 2 example 1 into the compound (3). As a result of observing change of fading after 72-hour exposure visually, without this resin moldings carrying out water injection with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

Except having changed the compound (1) in example 3 example 1 into the compound (6), the same actuation as an example 1 was repeated, and the master batch was obtained. They are ABS plastics (it mixed with the \*\*\*\* Naugatuck trade name "clara stick MH" 100 section, and the resin moldings was obtained by extrusion molding.) about this masterbatch 4 section. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

The same actuation as an example 3 was repeated except having replaced the compound (8) with for the compound (6) in example 4 example 3. As a result of observing change of fading after 100-hour exposure visually, without this resin moldings carrying out water injection with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted. Into example 5 flask, the melamine 10 section, the paraformaldehyde 60 section, and the p-toluenesulfonamide 40 section are taught, and melting is carried out at 50-100 degrees C. Heating stirring was performed for the compound (6) at 10 \*\*\*\*\* and further 120-150 degrees C for 30 minutes, and the melamine resin fluorescence coloring object was obtained. The powder of the fluorescence coloring agent which has clear yellow was obtained by a mortar's grinding this fluorescence coloring object lightly, and grinding with a ball mill further for 1 hour. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

The dioctyl phthalate 40 section and the stabilizer 5 section were added to the example 6 polyvinyl-chloride 100 section, and after mixing the fluorescence coloring agent obtained in the example 1 in the vinyl chloride powder 50 section processed with the conventional method to 1 \*\*\*\*\* homogeneity, respectively, the coloring chlorination vinyl sheet of kneading yellow was obtained for 10 minutes at 160 degrees C with 2 rolls. This coloring chlorination vinyl sheet was put on the mirror plane plate, press forming was carried out at the temperature of 160 degrees C using the thickness of 0.1mm, 100mm long, and a 100mm wide spacer, and the yellow coloured film-like Plastic solid was acquired. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, as for the fall of fluorescence, and change of coloring, 100 passage of time after was not accepted. The same actuation as an example 6 was repeated except having replaced the compound (1) with the compound (11) in example 7 example 6. As a result of observing change of fading after 100-hour exposure visually, without this fluorescence coloring agent carrying out water injection with a sunshine weather meter, as for the fall of fluorescence, and change of coloring, 100 passage of time after was not accepted.

Into the example 8 flask, the melamine 10 section, the paraformaldehyde 60 section, and the p-

toluenesulfonamide 40 section were taught, and melting was carried out at 50–100 degrees C. Heating stirring was performed for the compound (6) obtained in the example 2 at 10 \*\*\*\*\* and further 120–150 degrees C for 30 minutes, and the melamine resin coloring object was obtained. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

The same actuation as an example 8 was repeated except having replaced the compound (6) with the compound (10) in example 9 example 8. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

Into the example 10 flask, the methyl methacrylate 80 section, the divinylbenzene 20 section, and the azobisisobutyronitril 0.8 section were taught, and melting was carried out at 50–100 degrees C. The suspension of the fluorescence coloring object of a monomer which stirred the compound (9) to this by having added the polyvinyl alcohol 3 section and the water 200 section further 10 \*\*\*\*\*, and was colored yellow at it was obtained. This suspension was heated, the polymerization reaction was performed and polymerization hardening was carried out. The fluorescence coloring agent was separated from suspension and the powder of the fluorescence coloring agent which grinds with a ball mill for 1 hour, and has clear yellow was obtained after desiccation. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

The fluorescence coloring agent 5 section manufactured in the example 11 example 7, an alkyd resin distribution object [60% 55 sections of alkyd resin solid content, the melamine resin (50% of solid content) 35 section, the xylene 2 section, and the methyl glycol (2-methoxyethanol) 2 section], and the ceramic ball 230 section of 8mmphi were put into the 400ml glass jar, and it was made to distribute for 30 minutes in the rotational speed of 120rpm. The obtained distributed object was applied to the metal plate, it could be burned for 3 minutes at 180 degrees C, and the paint plate was obtained. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

The 1.0 sections, the varnish (styrene acrylic-acid type) 49 section for aquosity gravures, and the 3mmphi glass bead 150 section were put into the 225ml mayonnaise bottle for example 12 compound (2), the varnish 35 section for an addition was added after 90-minute distribution by the paint conditioner, the paint conditioner distributed for 10 more minutes, the glass bead was carried out the \*\* exception, and fluorescence aquosity rotogravure ink was obtained. As a result of measuring change of fading after 48-hour exposure with a color difference meter, without carrying out drawdown of this ink to the Manila board paper by #3 bar coating machine, and carrying out water injection for this with a sunshine weather meter and asking for Delta E, it was set to 2.5 and the fall of fluorescence and change of coloring were not accepted.

4 rotation kneading meat of the 0.5 sections and the varnish 50 section for rosin phenol resin system offset was carried out for example 13 compound (3) 150 lbs 100 revolutions by Hoover style Mahler, ink was adjusted and drawdown of this was carried out to art paper using the small rotary press (RI circuit tester). As a result of measuring change of fading after 48-hour exposure with a color difference meter, without carrying out water injection for this with a sunshine weather meter and asking for Delta E, it was set to 1.5 and the fall of fluorescence and change of coloring were not accepted.

Stirring mixing of the 0.3 sections, the petroleum-solvent (product made from shell chemistry, shell ZORU AB) 30.7 section, the dimethyl sulfoxide 15 section, and the N-methyl-2-pyrrolidone 54 section was carried out for 20 minutes, example 14 compound (3) was carried out the \*\* exception with the 0.45micro membrane filter, recording ink was made, and it recorded on the regular paper using the printer of a piezo method. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

1 N of examples of a comparison, the powder 30 section of N-di-n-butyl Quinacridone, polyethylene (trade name "SUMIKASEN G-808" by Sumitomo Chemical Co., Ltd. 30 section, and

polyethylene wax (the trade name by Mitsuhiro chemical-industry company "Sun Wacks 131P" 40 section was pelletized after kneading and with an extruder by the kneader, and the master batch was obtained.) It is high density polyethylene (it mixed with the trade name "high ZEKKUSU 2208" by Mitsui Petrochemical Industries, Ltd. 100 section, and the resin moldings was obtained by extrusion molding.) about this masterbatch 4 section. As a result of measuring change of fading after 48-hour exposure with a color difference meter, without carrying out water injection for this with a sunshine weather meter and asking for Delta E, it was set to 11.7 and the fall of fluorescence and change of coloring were accepted.

[0042] if the example 1 of a comparison is compared with an example 1, by introducing or more 4 organic residue into the basic frame of a general formula [1], lightfastness will be markedly alike and will improve -- it became clear.

[0043]

[Effect of the Invention] blending the fluorescence coloring agent of this invention with a plastics moldings, a coating constituent, an ink constituent, etc. -- a splendid fluorescence color is given and there is the description of moreover excelling in thermal resistance and lightfastness as compared with the fluorescence coloring agent known conventionally.

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TECHNICAL FIELD

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[Field of the Invention] This invention relates to the fluorescence coloring agent used for various plastics moldings, a coating, ink, etc. about a fluorescence coloring agent.

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PRIOR ART

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[Description of the Prior Art] Generally fluorescent dye is rich in reactivity, and inferior to the robustness over light. As the cure, there are an approach of scouring this in a formation object, and the approach of coating-izing and painting as coloring agent powder which dissolved fluorescent dye in synthetic resin. These coloring agents should not yet be satisfied, although weatherability improves and it has come to be equal to field use, when fluorescent dye is scoured in a direct formation object.

[0003] On the other hand, the condensed multi-ring system organic compound which makes Quinacridone representation has lightfastness, thermal resistance, and the property excellent in solvent resistance, and the coloring agent of various coatings, such as a coating for automobiles, and polymeric materials, printing ink, etc. are utilized widely. These condensed multi-ring system organic compounds are firmly combined by the stacking or the intermolecular hydrogen bridge, consequently it does not dissolve in an organic solvent, or it is poorly soluble very much, and generally fluorescence is not seen or only very weak fluorescence is observed.

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EFFECT OF THE INVENTION

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[Effect of the Invention] blending the fluorescence coloring agent of this invention with a plastics moldings, a coating constituent, an ink constituent, etc. -- a splendid fluorescence color is given and there is the description of moreover excelling in thermal resistance and lightfastness as compared with the fluorescence coloring agent known conventionally.

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TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention] Compatibility is high to a binder etc. and this invention aims at offering the fluorescence coloring agent excellent in lightfastness.

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## MEANS

[Means for Solving the Problem] It makes it easy for the fluorescence coloring agent shown in the general formula [1] to extend the intermolecular distance of a quinacridone pigment, and to distribute it by introducing four or more organic residue of carbon numbers 1-50 into the quinacridone pigment which was insoluble to the organic solvent until now, or its intermediate field. Consequently, when it was made the coating constituent which contains the binder for coatings when it is made the plastics moldings which it makes it come to distribute in plastics when making the fluorescence coloring agent of this invention contain in the resin of the shape of a particle which constructed the bridge, or when it was made the ink constituent containing an ink binder, pulling out the fluorescence which was not seen until now was found out.

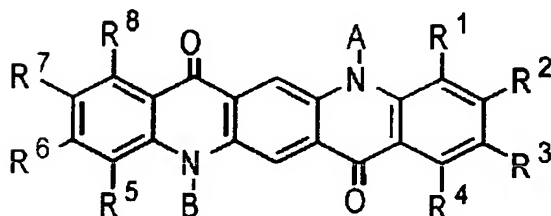
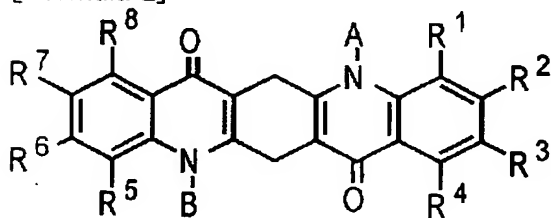
[0006] If concentration quenching explains this phenomenon, when the fused aromatic ring part of the Quinacridone molecule cannot approach mutually and will carry out it, molecule spacing is guessed for the phenomenon of breadth and quenching to stop being able to occur easily. When the fluorescence coloring agent shown in the general formula [1] introduced the organic residue of carbon numbers 1-50 into R1-R8, and A and B for the above reason, a stacking and an intermolecular hydrogen bridge are cut and it is expected to have become usable as a fluorescence coloring agent.

[0007] That is, this invention relates to the fluorescence coloring agent which consists of a compound shown by the following general formula [1].

General formula [1]

[0008]

[Formula 2]



[0009] [-- among a formula, although R1-R8, and A and B express a hydrogen atom or the organic residue of carbon numbers 1-50 independently, respectively, at least four of R1-R8, and A and B are the organic residue of 1 to 50 as for them.]

Furthermore, this invention relates to the above-mentioned fluorescence coloring agent with

which the melting point of the compound shown by the general formula [1] is characterized by being 250 degrees C or more.

[0010] Furthermore, this invention relates to the fluorescence coloring object which makes it come to contain the above-mentioned fluorescence coloring agent in the resin of the shape of a particle which constructed the bridge.

[0011] Furthermore, this invention relates to the fluorescence coloring plastic-molding object which makes it come to distribute the above-mentioned fluorescence coloring object in plastics.

[0012] Furthermore, this invention relates to the fluorescence coloring coating constituent containing the above-mentioned fluorescence coloring agent and the binder for coatings.

[0013] Furthermore, this invention relates to the fluorescence coloring ink constituent containing the above-mentioned fluorescence coloring agent and an ink binder.

[0014]

[Embodiment of the Invention] R1-R8 of the compound shown by the general formula [1] in this invention, and A and B are a hydrogen atom or the organic residue of carbon numbers 1-50, and the aryl thio radical which is not permuted [ the aryloxy group which is not permuted / the aryl group which is not permuted / the thio alkoxy group which is not permuted / the alkoxy group which be permuted / the alkyl group which be permuted / a permutation or / a permutation, or / a permutation, or / a permutation, or /, a permutation, or /, a permutation, or ] is mentioned as an example of the organic residue of carbon numbers 1-50.

[0015] As an alkyl group which is not permuted [ a permutation or ], a methyl group, an ethyl group, n-propyl group, An ISO-propyl group, butyl, sec-butyl, tert-butyl, A pentyl radical, a hexyl group, a heptyl radical, an octyl radical, the dodecyl, a tetradecyl radical, An EOSHIRU radical, a heptadecyl radical, a hexadecyl radical, a cyclohexyl radical, Others [ alkyl group / of the carbon numbers 1-50, such as an octadecyl radical and a stearyl radical, / non-permuted ], 2-phenyl isobutyl radical, a trifluoro pentyl radical, a hexafluoro pentyl radical, There is a permutation alkyl group of the carbon numbers 1-50, such as benzyl, alpha-phenoxy benzyl, alpha, and alpha-dimethylbenzyl radical, alpha, and alpha-methylphenyl benzyl, alpha, and alpha-ditrifluoromethyl benzyl, a triphenylmethyl radical, and alpha-benzyloxybenzyl radical.

[0016] As an alkoxy group which is not permuted [ a permutation or ] Others [ alkoxy group / of the carbon numbers 1-50, such as an n-butoxy radical, a t-butoxy radical, n-octyloxy radical, and t-octyloxy radical, / non-permuted ], There are permutation alkoxy groups of carbon numbers 1-50, such as 1, 1, and 1-tetrafluoro butoxy radical and a benzyloxy radical. As a thio alkoxy group which is not permuted [ a permutation or ] A thio n-butoxy radical, a thio-t-butoxy radical, a thio-n-octyloxy radical, There are permutation thio alkoxy groups of carbon numbers 1-50, such as 1 and 1, and 1-tetrafluoro thio butoxy radical and a benzyloxy radical. [ besides the non-permuted thio alkoxy group of the carbon numbers 1-50, such as a thio-t-octyloxy radical, ]

[0017] As an aryl group which is not permuted [ a permutation or ], there are a phenyl group, 2-methylphenyl radical, 3-methylphenyl radical, 4-methylphenyl radical, 4-ethyl phenyl group, a biphenyl radical, 4-methyl biphenyl radical, 4-ethyl biphenyl radical, 4-cyclohexyl biphenyl radical, a terphenyl radical, 3, 5-dichlorophenyl radical, a naphthyl group, 5-methyl naphthyl group, an anthryl radical, a pyrenyl radical, etc. As an aryloxy group which is not permuted [ a permutation or ], there are permutation aryloxy groups of carbon numbers 1-50, such as 1 and 1 besides the non-permuted aryloxy group of the carbon numbers 1-50, such as a phenoxy group, 4-ethyl phenoxy group, 4-propyl phenoxy group, a 4-n-butyl phenoxy group, a 4-t-butoxy phenoxy group, a 2-n-octyl phenoxy group, and a 3-t-octyl phenoxy group, 1-tetrafluoro phenoxy group, and a biphenyl oxy-radical.

[0018] As an aryl thio radical which is not permuted [ a permutation or ], there are a phenylthio radical, 2-methyl phenylthio radical, 3-methyl phenylthio radical, 4-methyl phenylthio radical, 4-ethyl phenylthio radical, a biphenyl thio radical, 4-methyl biphenyl thio radical, 4-ethyl biphenyl thio radical, 4-cyclohexyl biphenyl thio radical, a TAFENIOCHIO radical, 3, 5-dichloro phenylthio radical, a naphthyl thio radical, 5-methyl naphthyl thio radical, an anthryl thio radical, a pyrenyl thio radical, etc.

[0019] As an example of the substituent which organic residue may combine, there is an amino

group other than the above-mentioned functional group which is not permuted [ a halogen atom, a permutation, or ].

[0020] There are fluorine, chlorine, a bromine, and iodine as a halogen atom, and there are a phenyl methylamino radical besides dialkyl permutation amino groups, such as an amino group, a dimethylamino radical, and a diethylamino radical, a diphenylamino radical, a ditolylamino radical, a dibenzylamino radical, etc. as an amino group which is not permuted [ a permutation or ].

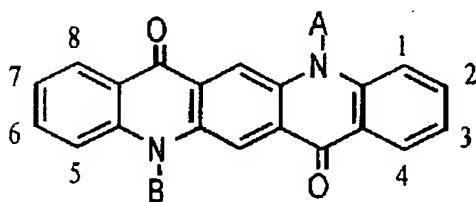
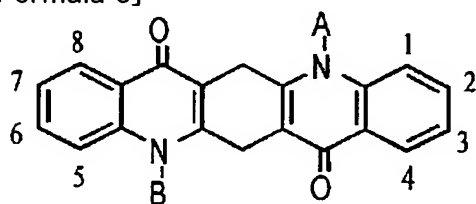
[0021] Moreover, by adjoining substituents, it may join together mutually, respectively and a phenyl group, a naphthyl group, an anthryl radical, a pyrenyl radical, etc. may be formed.

[0022] Especially the location that the organic residue of the compound of the general formula [1] in this invention may combine is not limited. However, it is desirable preferably that it is the 1.4.5.8th place shown in the general formula [2] or that A and B are organic residue, and when A or B is an aryl group, it is still more desirable that 1 or the organic big residue of steric hindrance is introduced the 5th place. It is thought that such organic residue serves to cut or pull apart an intermolecular hydrogen bridge.

General formula [2]

[0023]

[Formula 3]



[0024] [-- A and B express a hydrogen atom or the organic residue of carbon numbers 1-50 independently among a formula, respectively.]

generally, a fluorescent pigment or fluorescent dye has the low melting point, and it is fluorescent dye of versatility -- acid When it is red, it is 208 degrees C. When the melting point of a fluorescent pigment was low and it is made a masterbatch, the problem of discoloration, metal mold dirt, etc. is caused. In order to avoid these problems, the melting point of 250 degrees C or more is required at worst.

[0025] N,N-dibenzyl-1.5-dimethyl-Quinacridone which is the compound shown by the general formula [1]: The melting point of a compound (1) is as high as 350 degrees C, and it has strong fluorescence. The compound shown by the general formula [1] is compoundable by compounding Quinacridone using the raw material which introduces organic residue into a commercial quinacridone pigment and which is caused especially or has organic residue. The example of the compound shown by the general formula [1] below is illustrated to Table 1.

[0026]

[Table 1]

化合物	化学構造
(1)	
(2)	
(3)	

[0027]

化合物	化学構造
(4)	
(5)	
(6)	

[0028]

化合物	化学構造
(7)	
(8)	
(9)	

[0029]

化合物	化学構造
(10)	
(11)	
(12)	

[0030]

化合物	化学構造
(13)	
(14)	
(15)	

[0031]

化合物	化学構造
(16)	
(17)	
(18)	

[0032] In a solid state, it has strong fluorescence and the compound shown by the general formula [1] shows the high melting point. For this reason, although the compound shown by the general formula [1] can be used as a fluorescence coloring agent as it is, after distributing the compound shown by the general formula [1] in the resin of cross-linking, it can be made to be able to harden and what ground this to the particle can also be used as a fluorescence coloring agent. Moreover, after emulsion-izing the compound shown by the general formula [1] by the drainage system with aqueous resin using an emulsifier or a stabilizer, what was hardened and was made into the spherical particle can be used as a fluorescence coloring agent. As resin of the above-mentioned cross-linking, acrylic resin, styrene resin, aromatic series sulfonamide resin, an epoxy resin, an alkyd resin, a urea resin, melamine resin, benzoguanamine resin, acrylic urethane resin and those copolymers, copolycondensation resin, etc. can be mentioned. An ultraviolet ray absorbent, an anti-oxidant, a singlet oxygen quencher, a hindered amine light stabiliser, other stabilizers, and an additive can be blended with the above-mentioned resin.

[0033] 0.1-100 micrometers of mean particle diameter of said particle are 1-20 micrometers preferably. The concentration of the compound shown by the general formula [1] blended with the resin of a particle is 0.05 - 10 % of the weight preferably 0.01 to 20% of the weight. It becomes [ fluorescence intensity ] weak that it is low concentration and is not more desirable than 0.01 % of the weight.

[0034] As a more concrete manufacturing method of the fluorescence coloring agent which consists of particle-like resin For example, after adjusting the mixture of amino compounds, such as benzoguanamine and a melamine, and formalin so that pH may serve as the range of 5-10, In the primary reaction object of the amino resin which was made to react at the temperature of 50-100 degrees C, and was obtained The amino resin 100 weight section is received in the compound shown by the general formula [1]. 0.1 - 20 weight section, Under stirring in the water solution which carries out 0.5-10 weight section addition preferably, and contains protective colloid agents, such as polyvinyl alcohol, for this in the range of 1 - 30 weight section to the amino resin 100 weight section, It supplies and the suspension of amino resin is obtained, and subsequently, in addition, 40-100 degrees C performs polycondensation hardening for curing catalysts, such as a mineral acid and an organic acid, at this to the amino resin 100 weight section in the range of 0.01 - 10 weight section, and after it carries out the obtained hardening resin a \*\* exception and it carries out stoving, it crushes.

[0035] The fluorescence coloring agent which consists of the compound shown by the general formula [1] or a compound shown by the general formula [1], and bridge formation resin of a particle can be blended with shaping plastics. Under the present circumstances, the fluorescence coloring agent of this invention can be blended with high concentration, for example, extrusion molding can be carried out to the constituent which consists of dispersants, such as base resin and metal soap, and a wax, at a pellet type, and it can consider as a masterbatch. As resin of the tree for shaping plastics, there are polyolefin resin, such as polyethylene resin and polypropylene resin, polyester resin, polyamide resin, PVC resin, ABS plastics, styrene resin, acrylic resin, polycarbonate resin, urethane resin, amino resin, etc. Let these thermoplastics be a plastics moldings by the approach of inflation molding, calender shaping, and others. The fluorescence coloring agent of this invention can be blended with thermosetting plastic, such as an epoxy resin, phenol resin, polyester resin, polyester imide resin, and polyamidoimide resin.

[0036] Surfactants, such as rosin acid soap, stearin acid soap, oleic acid soap, Na-G beta-naphthyl methane JISARUFATO, Na-lauryl sulfate, Na-diethyl hexyl sulfo KUSHINETO, and Na-diocetyl sulfo KUSHINETO, can distribute, and the fluorescence coloring agent of this invention can be used as the coloring agent constituents a coating, for ink, etc.

[0037] The compound shown by the general formula [1] is meltable to an organic solvent, and can blend this with a coating or ink as remaining as it is or a fluorescence coloring agent of solvent insolubility which consists of a particle of resin. As ink, there are printing ink, such as offset-printing ink, gravure ink, and the silk screen, ink for ink jets, copying ink, etc. As paint resin and resin for ink, polymethyl acrylate, Acrylic resin, such as polyethylacrylate, polymethylmethacrylate, and polyvinyl acrylate, Polyurethane resin, acrylic urethane resin, polyester resin, polyarylate resin, Vinyl chloride resin, an ethylene-vinylacetate copolymer,

polyvinyl formal resin, Amino resin, an alkyd resin, an epoxy resin, phenol resin, polyester resin, Polyester imide resin, polyamidoimide resin, silicone resin, Polyvinylidene chloride resin, polyethylene vinyl alcohol resin, polyolefin resin, Elasticity, such as vinylidene chloride resin, butadiene resins, styrene resin, phenoxy resin, polyamide resin, cellophane, ethyl cellulose, and a nitrocellulose, rigid resin, etc. can be mentioned. The blending ratio of coal of the fluorescence coloring agent of this invention is 0.5 - 10 % of the weight preferably 0.1 to 20% of the weight to a coating constituent or the whole ink constituent.

[0038] The fluorescence coloring agent of this invention can raise light stability-proof by using it with an ultraviolet ray absorbent. An ultraviolet ray absorbent An o-hydroxy benzophenone, 2-hydroxy-4-n-octoxybenzophenone, Benzophenone systems, such as 2-hydroxy-4-methoxybenzophenone, 2-(2'-hydroxyphenyl) benzotriazol, 2-(2'-hydroxy-5'-t-octyl phenyl) benzotriazol, 2-(2'-hydroxy-3'-t-butyl -5'-methylphenyl)-5-chlorobenzo triazole, Benzotriazols, such as 2-(2'-hydroxy-5'-methylphenyl) benzotriazol, Ethyl-2-cyano - 3 and 3-diphenyl acrylate, 5-ethylhexyl-2-cyano - Cyanoacrylate systems, such as 3 and 3-diphenyl acrylate, Salicylic acid systems, such as phenyl salicylate and 4-t-butyl phenyl salicylate, Ultrafine particle inorganic systems, such as oxalic acid anilide systems, such as 2-ethyl-5'-t-butyl-2'-ethoxy-N and an N'-JIFENIROKISARU amide, a zinc oxide, titanium oxide, and a zirconium dioxide, etc. can be used.

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EXAMPLE

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[Example] Hereafter, this invention is further explained to a detail based on an example. The section expresses the weight section among an example.

The synthetic example 1 (composition of a compound (1))

Into the dioxane 300 section, the succinyl succinic-acid dimethyl 24 section, the 2-amino-1-methylbenzene 50 section, and the 35% hydrochloric-acid 1.5 section were put in, and heating churning was carried out at 95 degrees C for 3 hours. Then, it diluted with the methanol water of the 500 sections, and the rare sodium-hydroxide water solution neutralized. The depositing yellow solid-state was carried out the \*\*\*\*\* exception, and it washed in the methanol 500 section further. The obtained yellow solid-state was dried at 70 degrees C, and the needle crystal 43 section which has yellow fluorescence was obtained.

[0040] Heating stirring was performed for this yellow needle crystal 50 section at 250 degrees C under the nitrogen air current with the 1-methylnaphthalene 300 section for 10 hours. Then, the orange solid-state which added the methanol 300 section and deposited was carried out the \*\*\*\*\* exception. It washed in the methanol 300 section, the obtained orange solid-state was dried at 70 degrees C, and the light orange powder 28 section which has whitish and orange fluorescence was obtained.

[0041] Heating stirring was performed for this light orange crystal 20 section for 10 minutes at 280 degrees C under the air air current with the dibutyl phtalate 200 section. Then, the purple solid-state which added the methanol 300 section and deposited was carried out the \*\*\*\*\* exception. It washed in the methanol 300 section, the obtained purple solid-state was dried at 70 degrees C, and the purple powder 18 section which has orange fluorescence was obtained. Heating stirring was performed for this purple powder 18 section at 100 degrees C for 10 hours with the dichlorobenzene 400 section, the 50% caustic soda water-solution 1000 section, the benzyl triethyl ammonium chloride 9 section, and the benzyl bromide 160 section. Then, the chloroform 300 section was added and the resultant was extracted. This chloroform layer was washed with water until it became neutrality, and it condensed after desiccation, the column chromatography using silica gel refined, and the powder 8 section which reprecipitates with a methanol and has orange fluorescence was obtained. It checked that it was a compound (1) as a result of molecular weight analysis. The fluorescence spectrum of a product is shown in drawing 1. In addition, the melting point of this compound was 350 degrees C.

The synthetic example 2 (composition of a compound (3))

Heating churning of 1, the 5-dimethyl Quinacridone 8 section, the benzene 140 section, the 50% caustic soda water-solution 500 section, the benzyl triethyl ammonium chloride 6 section, and the alpha-BUROMO-O-xylene 50 section was violently carried out at 80 degrees C among the flask for 30 hours. Then, the decantation separated the solid-state part, the water of the 500 sections washed, and chloroform extracted. This chloroform layer was condensed and the solid-state 8 red section was obtained. The column chromatography using silica gel refined and the powder 3 section which reprecipitates with a methanol and has yellow fluorescence was obtained. It checked that it was a compound (3) as a result of molecular weight analysis. The fluorescence spectrum of a product is shown in drawing 2. In addition, this compound showed the melting point of 320 degrees C.

The powder 30 section of the compound (1) obtained in the example 1 of example 1 composition, polyethylene (trade name "SUMIKASEN G-808" by Sumitomo Chemical Co., Ltd. 30 section, and polyethylene wax (the trade name by Mitsuhiro chemical-industry company "Sun Wacks 131P" 40 section was pelletized after kneading and with an extruder by the kneader, and the master batch was obtained.) It is high density polyethylene (it mixed with the trade name "high ZEKKUSU 2208" by Mitsui Petrochemical Industries, Ltd. 100 section, and the resin moldings was obtained by extrusion molding.) about this masterbatch 4 section. As a result of observing change of fading after 72-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted. The same actuation as an example 1 was repeated except having changed the compound (1) in example 2 example 1 into the compound (3). As a result of observing change of fading after 72-hour exposure visually, without this resin moldings carrying out water injection with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted. Except having changed the compound (1) in example 3 example 1 into the compound (6), the same actuation as an example 1 was repeated, and the master batch was obtained. They are ABS plastics (it mixed with the \*\*\*\* Naugatuck trade name "clara stick MH" 100 section, and the resin moldings was obtained by extrusion molding.) about this masterbatch 4 section. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

The same actuation as an example 3 was repeated except having replaced the compound (8) with for the compound (6) in example 4 example 3. As a result of observing change of fading after 100-hour exposure visually, without this resin moldings carrying out water injection with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted. Into example 5 flask, the melamine 10 section, the paraformaldehyde 60 section, and the p-toluenesulfonamide 40 section are taught, and melting is carried out at 50-100 degrees C. Heating stirring was performed for the compound (6) at 10 \*\*\*\*\* and further 120-150 degrees C for 30 minutes, and the melamine resin fluorescence coloring object was obtained. The powder of the fluorescence coloring agent which has clear yellow was obtained by a mortar's grinding this fluorescence coloring object lightly, and grinding with a ball mill further for 1 hour. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

The dioctyl phthalate 40 section and the stabilizer 5 section were added to the example 6 polyvinyl-chloride 100 section, and after mixing the fluorescence coloring agent obtained in the example 1 in the vinyl chloride powder 50 section processed with the conventional method to 1 \*\*\*\*\* homogeneity, respectively, the coloring chlorination vinyl sheet of kneading yellow was obtained for 10 minutes at 160 degrees C with 2 rolls. This coloring chlorination vinyl sheet was put on the mirror plane plate, press forming was carried out at the temperature of 160 degrees C using the thickness of 0.1mm, 100mm long, and a 100mm wide spacer, and the yellow coloured film-like Plastic solid was acquired. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, as for the fall of fluorescence, and change of coloring, 100 passage of time after was not accepted. The same actuation as an example 6 was repeated except having replaced the compound (1) with the compound (11) in example 7 example 6. As a result of observing change of fading after 100-hour exposure visually, without this fluorescence coloring agent carrying out water injection with a sunshine weather meter, as for the fall of fluorescence, and change of coloring, 100 passage of time after was not accepted.

Into the example 8 flask, the melamine 10 section, the paraformaldehyde 60 section, and the p-toluenesulfonamide 40 section were taught, and melting was carried out at 50-100 degrees C. Heating stirring was performed for the compound (6) obtained in the example 2 at 10 \*\*\*\*\* and further 120-150 degrees C for 30 minutes, and the melamine resin coloring object was obtained. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

The same actuation as an example 8 was repeated except having replaced the compound (6)

with the compound (10) in example 9 example 8. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

Into the example 10 flask, the methyl methacrylate 80 section, the divinylbenzene 20 section, and the azobisisobutyronitril 0.8 section were taught, and melting was carried out at 50-100 degrees C. The suspension of the fluorescence coloring object of a monomer which stirred the compound (9) to this by having added the polyvinyl alcohol 3 section and the water 200 section further 10 \*\*\*\*\*, and was colored yellow at it was obtained. This suspension was heated, the polymerization reaction was performed and polymerization hardening was carried out. The fluorescence coloring agent was separated from suspension and the powder of the fluorescence coloring agent which grinds with a ball mill for 1 hour, and has clear yellow was obtained after desiccation. this fluorescence coloring agent -- 108-degree C constant temperature -- even if saved in the layer for 2 hours, the fall of fluorescence and change of coloring were not accepted.

The fluorescence coloring agent 5 section manufactured in the example 11 example 7, an alkyd resin distribution object [60% 55 sections of alkyd resin solid content, the melamine resin (50% of solid content) 35 section, the xylene 2 section, and the methyl glycol (2-methoxyethanol) 2 section], and the ceramic ball 230 section of 8mmphi were put into the 400ml glass jar, and it was made to distribute for 30 minutes in the rotational speed of 120rpm. The obtained distributed object was applied to the metal plate, it could be burned for 3 minutes at 180 degrees C, and the paint plate was obtained. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

The 1.0 sections, the varnish (styrene acrylic-acid type) 49 section for aquosity gravures, and the 3mmphi glass bead 150 section were put into the 225ml mayonnaise bottle for example 12 compound (2), the varnish 35 section for an addition was added after 90-minute distribution by the paint conditioner, the paint conditioner distributed for 10 more minutes, the glass bead was carried out the \*\* exception, and fluorescence aquosity rotogravure ink was obtained. As a result of measuring change of fading after 48-hour exposure with a color difference meter, without carrying out drawdown of this ink to the Manila board paper by #3 bar coating machine, and carrying out water injection for this with a sunshine weather meter and asking for Delta E, it was set to 2.5 and the fall of fluorescence and change of coloring were not accepted.

4 rotation kneading meat of the 0.5 sections and the varnish 50 section for rosin phenol resin system offset was carried out for example 13 compound (3) 150 lbs 100 revolutions by Hoover style Mahler, ink was adjusted and drawdown of this was carried out to art paper using the small rotary press (RI circuit tester). As a result of measuring change of fading after 48-hour exposure with a color difference meter, without carrying out water injection for this with a sunshine weather meter and asking for Delta E, it was set to 1.5 and the fall of fluorescence and change of coloring were not accepted.

Stirring mixing of the 0.3 sections, the petroleum-solvent (product made from shell chemistry, shell ZORU AB) 30.7 section, the dimethyl sulfoxide 15 section, and the N-methyl-2-pyrrolidone 54 section was carried out for 20 minutes, example 14 compound (3) was carried out the \*\* exception with the 0.45micro membrane filter, recording ink was made, and it recorded on the regular paper using the printer of a piezo method. As a result of observing change of fading after 100-hour exposure visually, without carrying out water injection for this with a sunshine weather meter, the fall of fluorescence and change of coloring were not accepted.

1 N of examples of a comparison, the powder 30 section of N-di-n-butyl Quinacridone, polyethylene (trade name "SUMIKASEN G-808" by Sumitomo Chemical Co., Ltd. 30 section, and polyethylene wax (the trade name by Mitsuhiro chemical-industry company "Sun Wacks 131P" 40 section was pelletized after kneading and with an extruder by the kneader, and the master batch was obtained.) It is high density polyethylene (it mixed with the trade name "high ZEKKUSU 2208" by Mitsui Petrochemical Industries, Ltd. 100 section, and the resin moldings was obtained by extrusion molding.) about this masterbatch 4 section. As a result of measuring change of fading after 48-hour exposure with a color difference meter, without carrying out water injection

for this with a sunshine weather meter and asking for Delta E, it was set to 11.7 and the fall of fluorescence and change of coloring were accepted.

[0042] if the example 1 of a comparison is compared with an example 1, by introducing or more 4 organic residue into the basic frame of a general formula [1], lightfastness will be markedly alike and will improve -- it became clear.

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[Translation done.]

\* NOTICES \*

JP0 and NCIP1 are not responsible for any damages caused by the use of this translation.

1.This document has been translated by computer. So the translation may not reflect the original precisely.

2.\*\*\*\* shows the word which can not be translated.

3.In the drawings, any words are not translated.

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DESCRIPTION OF DRAWINGS

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[Brief Description of the Drawings]

[Drawing 1] The fluorescence spectrum of a compound (1)

[Drawing 2] The fluorescence spectrum of a compound (3)

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[Translation done.]

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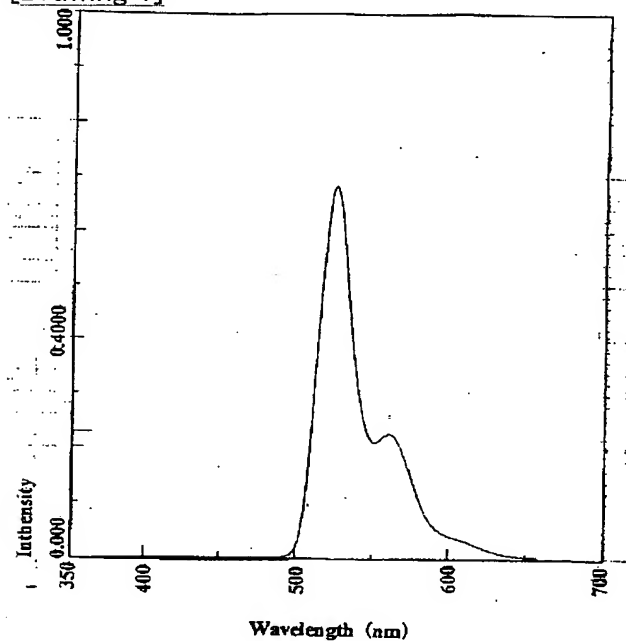
- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
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- 3.In the drawings, any words are not translated.

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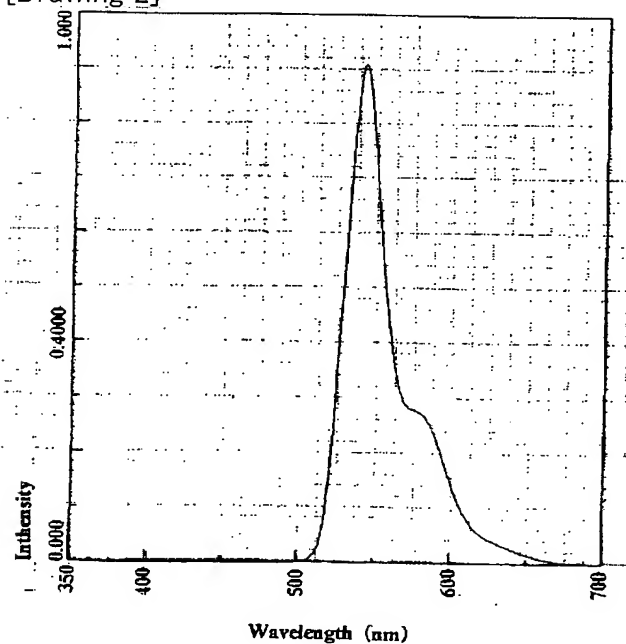
DRAWINGS

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[Drawing 1]



[Drawing 2]



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[Translation done.]